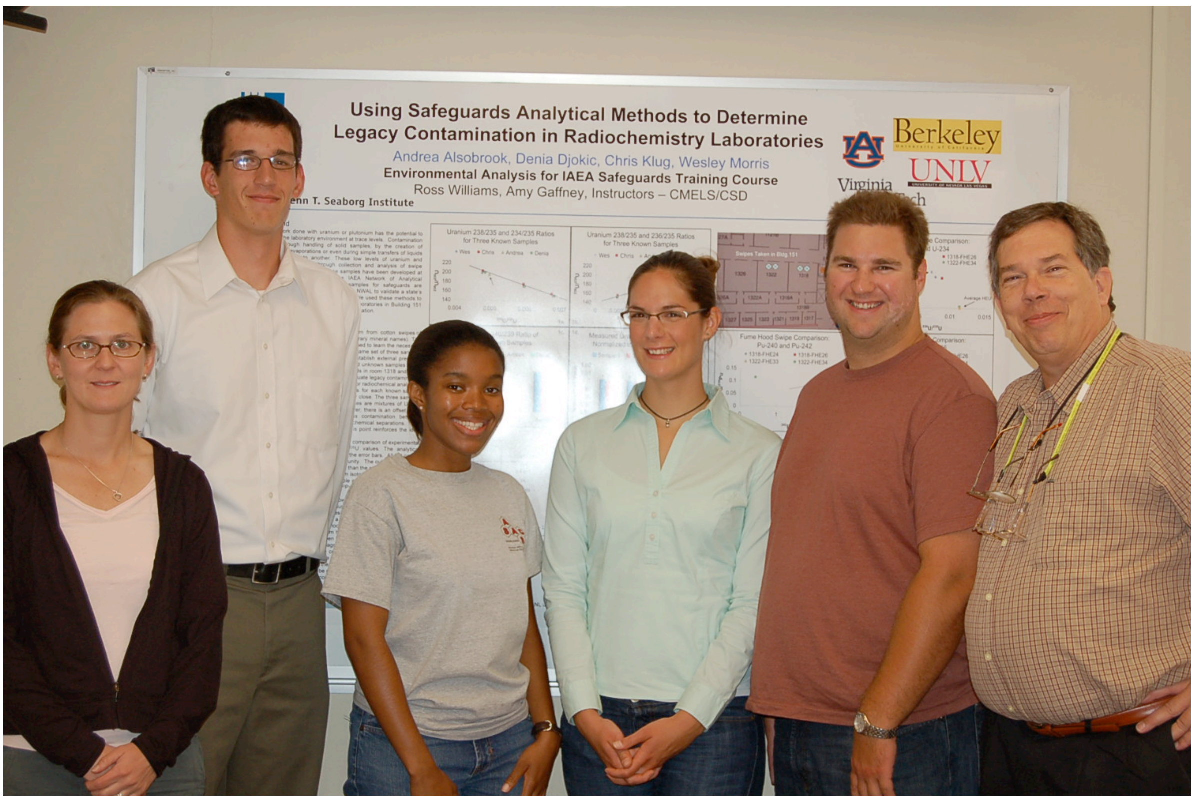
2008 LLNL Training in Environmental Sample Analysis for IAEA Safeguards

Lawrence Livermore National Laboratory
Livermore, CA 94550, USA

Instructors: Ross Williams, staff scientist, LLNL, Amy Gaffney, post-doctoral researcher, LLNL



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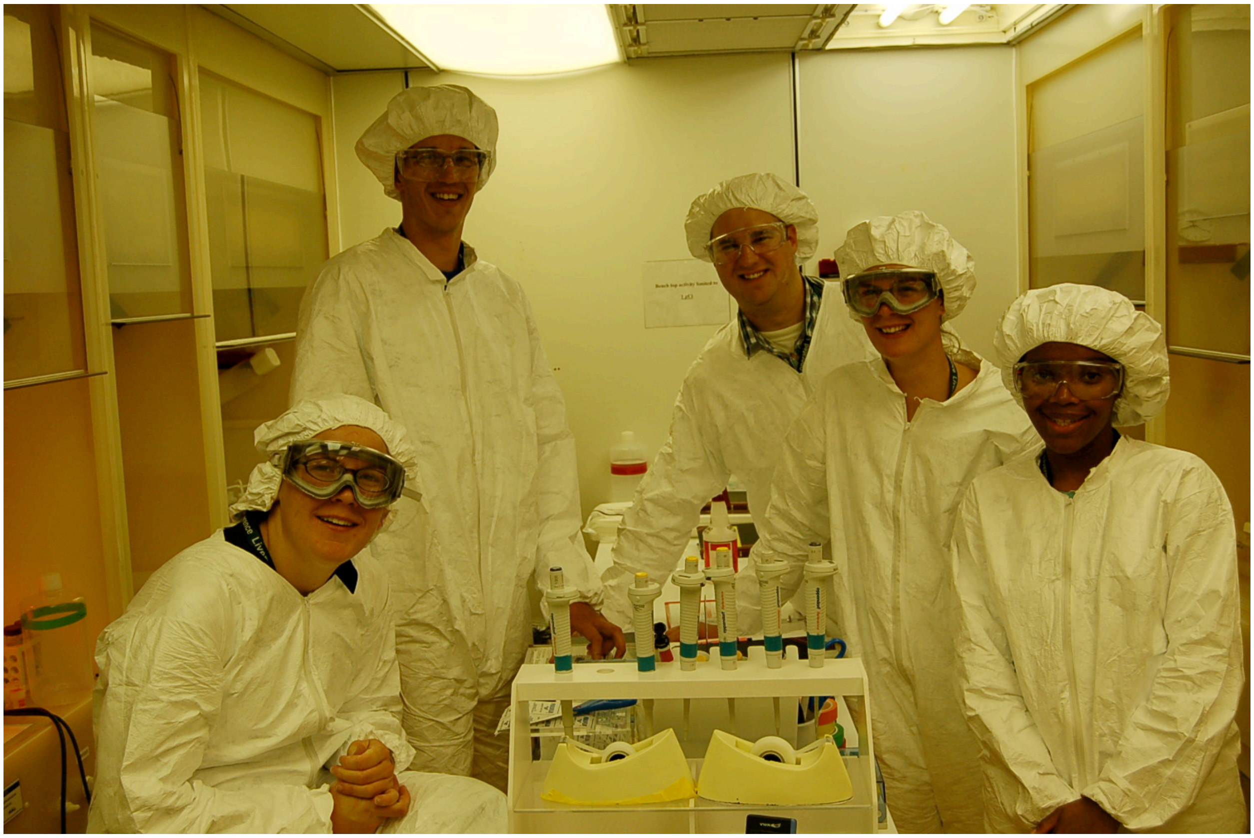
LLNL-AR-407087

A new course, Training in Environmental Sample Analysis for IAEA Safeguards, was offered this summer in the Chemistry, Materials, Earth and Life Sciences Directorate of LLNL. The course was taught by Ross Williams, staff scientist in the Environmental Radiochemistry group of CMELS, and Amy Gaffney, post-doctoral researcher. During the 8 week program, five students (four graduate students and one undergrad), from universities across the US, were trained in the analysis of bulk environmental samples for safeguards through hands-on experience working in a clean laboratory, purifying U and Pu from bulk environmental samples, and measuring U and Pu isotope ratios by multi-collector ICP mass spectrometry. The students made analyses of QA/QC samples prepared at LLNL for the DOE/NWAL and for other international round-robin comparisons. A series of lectures by invited safeguards and non-proliferation experts, gave the students a broad picture of the safeguards work of the IAEA. At the end of the course, the students prepared a poster of their work to showcase at LLNL's summer student poster symposium.

The program was developed in response to the recommendation in *International Safeguards Challenges and Opportunities for the 21st Century* that DOE/NNSA and the national laboratories develop a program to encourage experts to pursue IAEA careers. The students were recruited and hired as paid interns under the aegis of the Glenn T. Seaborg Institute (GTSI) at LLNL. The GTSI serves as a national center for the education and training of the next generation of scientists in the fields of nuclear chemistry, chemical engineering, materials science, environmental chemistry and chemical biology.



Speaker Schedule 2008

June 26	Ken Moody, LLNL	Forensic Radiochemistry
July 3	George Anzelon, LLNL	IAEA Investigations of Undeclared Nuclear Activities
July 10	Bill Dunlop, LLNL	Nuclear Weapons 101
July 17	John Perkins, LLNL	Fusion Energy
July 22	George Anzelon, LLNL	Uranium Enrichment and International Safeguards
July 24	Jean Moran, LLNL	Forensic Hydrology
July 31	Jay Davis, former director of DTRA	A Functional Look at the Nuclear Force
Aug 5	Ian Hutcheon	Nuclear Forensics: The Science and Some Real World Examples

Class Participants

Student	**Major**	**University**	**Year**
Andrea Alsobrook	Radiochemistry	Auburn University	Grad
Denia Djokic	Nuclear Engineering	UC Berkeley	Grad
Kathryn Flynn	Geochemistry	UC Davis	Grad
Chris Klug	Radiochemistry	U Nevada, Las Vegas	Grad
Wesley Morris	Chemistry	Vriginia Tech	Undergrad

Training in Environmental Sample Analysis for IAEA Safeguards

Syllabus

Week 1 June 17-20:
Introduction to LLNL. Training, training, and training.
Independent Study: Find some published methods for U and Pu purification from environmental samples and compare these with the LLNL NWAL procedure (copy provided).

Week 2 June 23-27
Group Meeting (June 24)
The IAEA NWAL. What is it? Why make "Analyses of Low-Level U and Pu in Bulk Environmental Samples" ?

Working in a clean laboratory: contamination control and good laboratory practices. Observe LLNL NWAL method in practice.
Establish lab use schedule: Who, where, when. Finish training and get ready to do chemistry.
Begin analysis of QC swipes.

Practice problems: Isotope ratios and the atomic mass of U.

Week 3 June 30-July3:
Group Meeting (July 1)
Inorganic isotope ratio mass spectrometry – Part 1: The fundamentals

Supervised laboratory work. Group mass spectrometry and instrument training sessions. An overview of MS data collection and analysis.

Week 4 July 7-11:
Group Meeting (July 8)
Inorganic isotope ratio mass spectrometry – Part 2: Ion-counting and statistics

Individual mass spectrometry instrument analysis sessions – uranium.
Finish purifications of U and Pu from QC swipe samples.
Begin work on unknown swipe sample from B151.

Practice problem: Spike calibration.

Week 5 July 14-18:
Group Meeting (July 15)
Gamma-ray spectrometry – an introduction and its role in safeguards
Guide to the Expression of Uncertainty in Measurement (GUM)

Individual mass spectrometry instrument analysis sessions – plutonium.
Finish chemistry on unknown swipe.

Hands-on with GUM: problems to solve.

Week 6 July 21-25:
Group meeting (July 22)
U-enrichment, reactor operations and interpretation of data from bulk environmental samples

Finish laboratory work and instrument analytical sessions.

Week 7 July 28-Aug.1:
Group meeting (July 29)
Nuclear forensics and international safeguards

Complete data analysis and compare results for the QC samples with the reference values. Laboratory work and instrument analyses are completed; lessons learned.

Week 8 August 4-8:
Last group meeting (Aug 5)

Interpret data and prepare poster for poster session. Poster must be uploaded to Information Management system for internal review no later than August 8; sooner is better. Start working on project report.

Week 9 August 11-15:
No group meeting.

Finish writing short project report. Print poster for poster session. Provide feedback and evaluations for this summer training program.

August 14: Present results in LLNL-wide poster session for summer intern

Auspices

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Using Safeguards Analytical Methods to Determine Legacy Contamination in Radiochemistry Laboratories

Andrea Alsobrook, Denia Djokic, Chris Klug, Wesley Morris

Environmental Analysis for IAEA Safeguards Training Course

Ross Williams, Amy Gaffney, Instructors – CMELS/CSD

Glenn T. Seaborg Institute

Background

Any work done with uranium or plutonium has the potential to contaminate the laboratory environment at trace levels. Contamination may occur through handling of solid samples, by the creation of aerosols during evaporations or even during simple transfers of liquids from one container to another. These low levels of uranium and plutonium are monitored through collection and analysis of swipe samples. Methods to analyze these samples have been developed at LLNL which is a member of the IAEA Network of Analytical Laboratories (NWAL). Environmental samples for safeguards are collected by the IAEA and analyzed by the NWAL to validate a state's declaration of activity at a nuclear facility. We used these methods to analyze environmental swipes taken from laboratories in Building 151 to investigate the presence of legacy contamination.

Results and Discussion

We separated uranium and plutonium from cotton swipes of known samples (listed below-right with arbitrary mineral names). The analyses of the known samples were performed to learn the necessary methods and demonstrate proficiency. The same set of three samples was analyzed by each summer intern to establish external precision limits for these techniques. We also analyzed unknown samples taken as swipes from the dust on top of fume hoods in room 1318 and 1322 of Building 151. These data are used to evaluate legacy contamination in these laboratories with a long use history for radiochemical analysis.

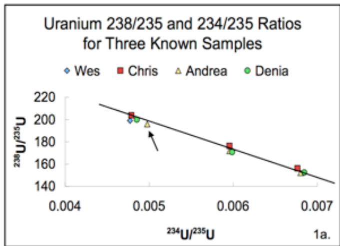
Figures 1a and 1b show that results for each known sample measured by different students are relatively close. The three samples define a line, demonstrating that the samples are mixtures of U with two different isotopic compositions. However, there is an offset point (black arrow) that possibly reflects cross contamination between samples, which probably occurred during chemical separations. The preservation of the linear trend through this point reinforces the idea that two samples were mixed.

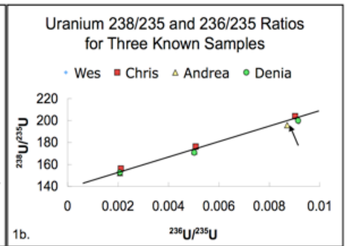
The bar graph (Figure 1d) shows a comparison of experimental results normalized to the known $^{238}U/^{235}U$ values. The analytical uncertainty for each analysis is shown by the error bars. All but one of the error bars on the results overlap with unity. The outlier indicated in Figure 1b has a $^{238}U/^{235}U$ value 2% lower than the others.

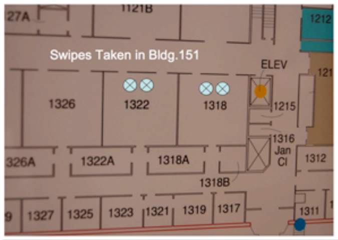
Figure 1c represents the plutonium isotope ratios. The height of the bars for $^{240}Pu/^{239}Pu$ from each sample are tightly grouped showing a relatively good agreement between $^{240}Pu/^{239}Pu$ measurements made by different analysts.

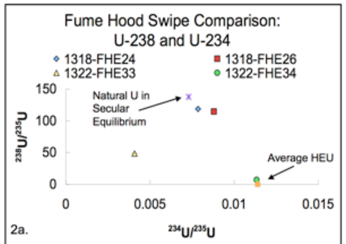
Figures 2a, 2b, and 2c show the isotopic composition of environmental samples taken from dust accumulated on top of fume hoods (FHE) in the sampled labs. Results indicate an enriched level of U-235 (12%) on FHE34, as well as a high ratio of Pu-242 to other Pu isotopes on the three other fume hoods (FHEs 24, 26, 33). We could conclude that low-enriched uranium has been handled in Room 1322, whereas Pu-242 tracers have been handled in both rooms. The linear trend for three points on the Pu diagram suggests that only two Pu end-members were sampled, but the complex use history of these rooms raises the possibility of many more components.

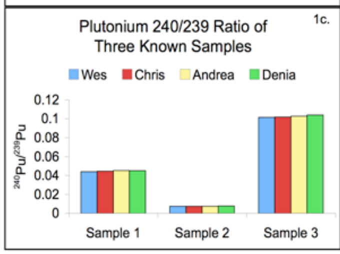
It should be noted that the isotopes detected by this extremely sensitive technique would not be detectable by common radioanalytical methods, and are well below acceptable levels.

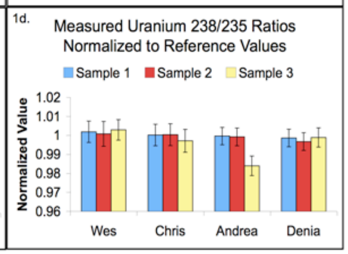


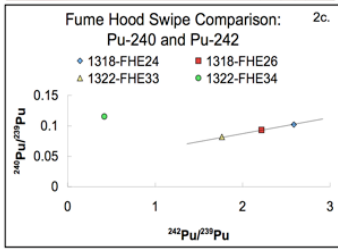


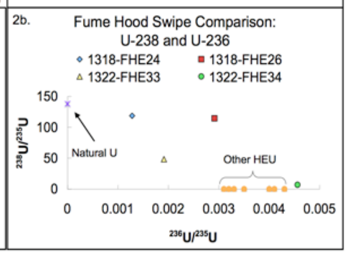












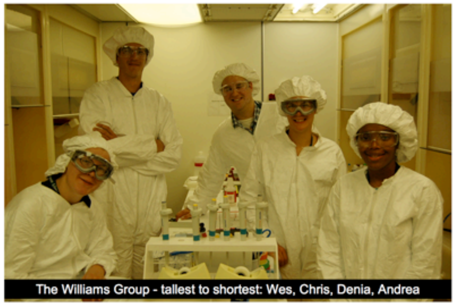
Reference Uranium Enrichment Ratios	U-238/U-235
90% - Weapons Grade	0.1111
15% - Low Enriched	5.6667
4% - Typical LWR Enrichment Level	24
Natural Uranium	137.88

Typical Pu Ratios for Pu from different sources	Pu-240/Pu-239	Pu-242/Pu-239
Light Water Reactor with typical 42 GWd/t burnup	0.47	0.09
Weapons Grade	0.06	0.01

Known Samples		mass of U on swipe (ng)	mass of Pu on swipe (pg)
WES	Witherite	127.53	59.12
	Wollastonite	118.50	59.45
	Wurtzite	121.61	61.68
CHRIS	Calcite	127.28	61.34
	Clinoenstatite	121.88	61.33
	Celestine	117.76	62.20
ANDREA	Aragonite	103.82	55.80
	Aegirine	129.13	59.72
	Anhydrite	124.86	110.11
DENIA	Dolomite	142.95	62.99
	Digenite	131.40	61.33
	Diopside	136.89	62.08

Fume Hood Unknowns		mass of U on swipe (ng)	mass of Pu on swipe (pg)
ANDREA	1318-FHE24	111.35	0.46
	1318-FHE26	52.76	0.34
WES	1322-FHE33	121.61	0.64
DENIA	1322-FHE34	71.90	1.09

Swipe samples of dust from the top of the hood



The Williams Group - tallest to shortest: Wes, Chris, Denia, Andrea

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Post #